

Supplementary information

The dripping-to-jetting transition in a co-axial flow of aqueous two-phase systems with low interfacial tension

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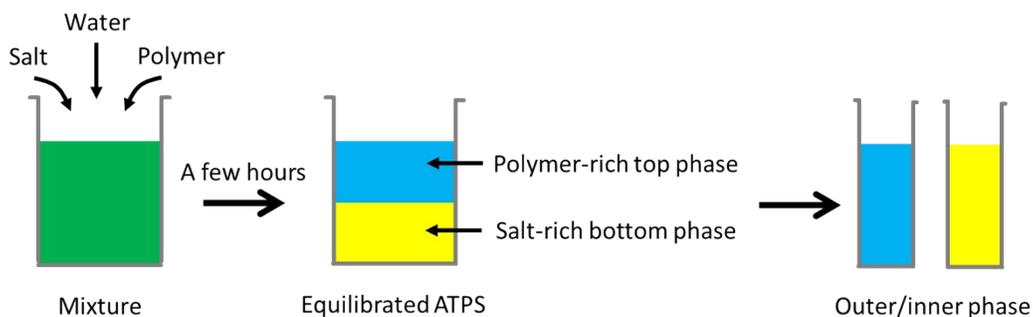
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1. Preparation of aqueous two-phase systems (ATPSs)
2. Compositions and fluid properties of ATPSs
3. Movies of dripping and jetting morphologies
4. Phase map of We_i and Ca_o for ATPSs
5. Expression for estimation of interfacial tension

1. Preparation of aqueous two-phase systems (ATPSs)

Known amount of salt, polymer and water are mixed. A sufficient time (several hours) is allowed for complete partitioning of the two immiscible aqueous phases. The polymer-rich top phase and the salt-rich bottom phase are collected for injection as the inner and outer phase in the dripping-to-jetting transition experiments.



SUPP FIG 1. Preparation of equilibrated ATPS

2. Compositions and fluid properties of ATPSs

Immiscible aqueous phases are prepared by dissolving and phase separating high concentrations of a polymer, Poly(Ethylene Glycol) 4000, 8000 (10 wt% to 23.29 wt%), and a salt, sodium citrate (10 wt% to 12 wt%) or sodium carbonate (6.44 wt% to 10.27 wt%) into deionized water. The specific compositions and corresponding fluid properties of the ATPSs and oil-water system are listed in Table S1. The viscosity ratio μ_i/μ_o varies from 0.04 to 45. The interfacial tension and viscosity are measured using a spinning drop tensiometer (SITE 100, Krüss) and microfluidic viscometer (MicroVisc, RheoSense) respectively. For all the ATPSs, the top phase is more viscous than the bottom phase. For the oil-water system, we use deionized water as the outer phase and silicone oil (PDMS 1 cSt) as the inner phase.

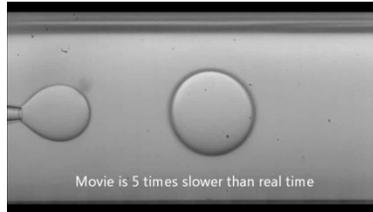
SUPP TABLE 1. Compositions and fluid properties of ATPSs. Two immiscible aqueous phases were obtained by phase separation of the mixture with compositions listed in the first column. Subscripts T and B refer to the top and bottom phases respectively.

Compositions of the mixture (wt%)	γ (mN/m)	ρ_T (kg/m ³)	ρ_B (kg/m ³)	μ_T (mPa·s)	μ_B (mPa·s)	μ_B/μ_T
10% PEG 8000 + 10% Sodium citrate	0.02	1056	1111	13	8.4	0.67
10% PEG 8000 + 12% Sodium citrate	0.03	1081	1110	10	2.5	0.25
14.04% PEG 4000 + 6.44% Sodium carbonate	0.26	1072	1092	16	1.5	0.09
18.9% PEG 4000 + 7.945% Sodium carbonate	0.99	1086	1141	47	2.2	0.05
23.285% PEG 4000 + 9.7% Sodium carbonate	2.55	1087	1189	83	3.3	0.04
19% PEG 4000 + 16% Tripotassium phosphate	2.14	1096	1250	96	2.2	0.02

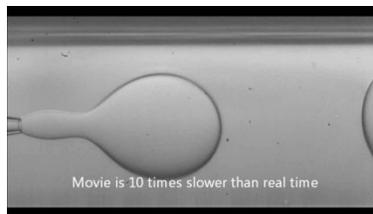
3. Movies of dripping and jetting morphologies

Transition to a widening jet by increasing the flow rate of the inner:

$$\gamma = 0.26 \text{ mN/m}, \mu_i = 1.5 \text{ mPa}\cdot\text{s}, Q_o = 500 \text{ }\mu\text{l/h.}$$



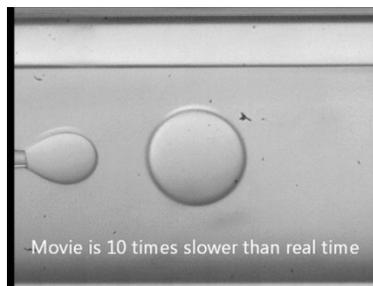
MOVIE S1. Dripping at $Q_i = 30 \text{ }\mu\text{l/h.}$



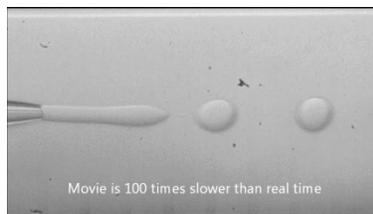
MOVIE S2. Jetting at $Q_i = 100 \text{ }\mu\text{l/h.}$

Transition to a narrowing jet by increasing the flow rate of the outer phase:

$$\gamma = 0.99 \text{ mN/m}, \mu_o = 47 \text{ mPa}\cdot\text{s}, Q_i = 100 \text{ }\mu\text{l/h.}$$



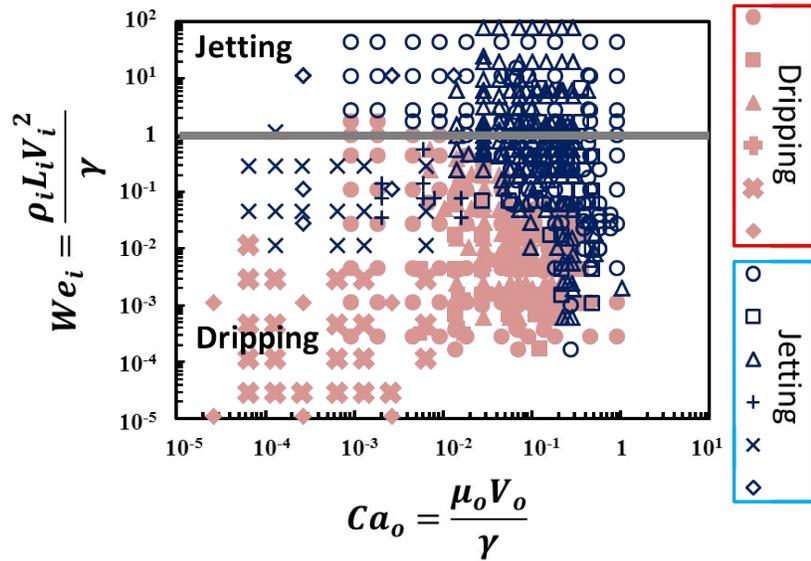
MOVIE S3. Dripping at $Q_o = 1 \text{ ml/h.}$



MOVIE S4. Jetting at $Q_o = 8 \text{ ml/h.}$

4. Figure of We_i against Ca_o for ATPSs

The dripping-to-jetting transition typically takes place at $We_i \sim 1$ for high interfacial tension and low viscosity. However, when the interfacial tension is low, as in the case of ATPSs, the Weber number at the transition can be one or two decades lower than unity. Hence, Weber number is not the appropriate indicator of the transition when the interfacial tension is low.



SUPP FIG S2. We_i against Ca_o for different ATPSs. Dripping and jetting are denoted by red and blue symbols. The grey solid line is $We_i \sim 1$. Circle: $\mu_i/\mu_o = 0.04$, $\gamma = 2.6$ mN/m. Square: $\mu_i/\mu_o = 0.05$, $\gamma = 0.99$ mN/m; Triangle: $\mu_i/\mu_o = 0.09$, $\gamma = 0.26$ mN/m. Plus: $\mu_i/\mu_o = 10.6$, $\gamma = 0.26$ mN/m; Cross: $\mu_i/\mu_o = 25.2$, $\gamma = 2.6$ mN/m; Diamond: $\mu_i/\mu_o = 44.7$, $\gamma = 2.1$ mN/m.

5. Expression for estimation of interfacial tension

The dripping-to-jetting transition occurs when the term $\frac{\sum F_i}{F_{IFT}} + \frac{\sum F_o}{F_{IFT}}$ lies between 0.1 and 1. This term is inversely proportional to the interfacial tension γ . Hence the interfacial tension γ should be estimated based on a multiplying factor ranging from 1 to 10, reciprocals of 1 and 0.1 respectively. In this work, a multiplying factor of 5, which is halfway between 1 and 10, has been chosen.